Enhanced Control of Mercury and Other HAPs by Innovative Modifications to Wet FGD Processes

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Introduction

The 1990 Clean Air Act Amendments mandated the Environmental Protection Agency to study the health effects caused by hazardous air pollutants (HAPs) from electric utility plants. The act also mandated a separate study on the effect of mercury emissions. Most HAPs of concern in power plants occur in the particulate phase and are therefore removed in particulate control devices. However, mercury, although emitted in extremely low concentrations, is primarily present in the vapor phase at most plants. Therefore, particulate removal devices are generally not effective at removing mercury from flue gas and alternative removal methods are needed. A number of different research programs have focused on the study of mercury emissions from power plants and methods for reducing these emissions.

The U.S. Department of Energy's Federal Energy Technology Center (DOE/FETC) is co-funding this project to further investigate a process for improving the ability of existing wet flue gas desulfurization (FGD) systems to control mercury emissions from coal-fired power plants. The project is being conducted under a cost-sharing PRDA agreement between DOE/FETC, Radian

International, and EPRI. The contract number is DE-AC22-95PC95260, and the period of performance extends through January 31, 2000. Tom Brown is the DOE/FETC Contracting Officer's Representative.

The proposed process consists of using a catalyst material to oxidize elemental mercury to a water-soluble mercury form. Vapor-phase mercury generally exists in two forms in utility flue gas—as elemental mercury and as water soluble, oxidized mercury (the predominant form is believed to be HgCl₂). Previous test results have shown that wet scrubbers effectively remove oxidized mercury from the gas but are ineffective in removing elemental mercury. Since elemental mercury is present in most flue gas streams, this process can potentially improve overall mercury removal in wet scrubbers by converting the elemental mercury to a removable form. During Phase I of this PRDA project, several catalyst materials were identified in the laboratory and in short-term field tests as being able to oxidize elemental mercury. Phase II of this project recently began, and will further investigate this process by exposing selected catalyst materials to flue gas over an extended period of time at various coal-fired power plants. This paper presents a summary of Phase I, the proposed Phase II testing approach, and preliminary Phase II results.

Objectives

The overall objective of this project is to learn more about controlling emissions of hazardous air pollutants (HAPs) from coal-fired power plants that are equipped with wet FGD systems. This objective is being pursued through two separate phases of a Mega-PRDA test program. Phase I of this project was completed in September 1997 and focused on three research areas:

- Catalytic oxidation of vapor-phase elemental mercury;
- Enhanced particulate-phase HAPs removal by electrostatic charging of liquid droplets; and
- Enhanced mercury removal by addition of additives to FGD process liquor.

Of these three areas, catalytic oxidation of elemental mercury showed the most promise as a potential commercial process for improving overall mercury removal. Based on promising results from Phase I, DOE/FETC selected this project for further investigation in a Phase II effort. The objectives of Phase II include:

- Estimate the ability of catalyst materials identified in Phase I to oxidize elemental mercury at various full-scale facilities;
- Estimate the life of these catalyst materials in various flue gas streams; and
- Estimate the volume of catalyst required to achieve at least 70% oxidation of the elemental mercury.

Approach

The concept of using a catalyst to oxidize vapor-phase elemental mercury was initially investigated at three scales of testing during Phase I of the PRDA. First, bench-scale tests were conducted to screen potential catalyst and fly ash materials for catalytic activity. Based on the results from these tests, the most promising catalysts were tested at EPRI's Environmental Control Technology Center in Barker, New York, using a 4-MW pilot FGD system. Following the pilot tests, additional catalyst testing was conducted at a lignite-fired utility power plant.

Phase I results from bench-scale, pilot-scale, and field tests indicated that several materials could oxidize elemental mercury to a chemical form more easily removed in wet scrubbers. To further develop this process, Phase II testing will address two critical issues – the life of the catalyst (i.e., how long will the catalyst oxidize mercury?) and the applicability of the process for the U.S. electric utility industry (i.e., are there coals/plants for which the catalyst does not work?). The catalyst oxidation efficiency and life will ultimately affect the form of the commercial process and its economics. The Phase II test program has been designed to address these issues by performing long-term catalyst tests at three full-scale utility sites.

Figure 1 illustrates the catalyst test unit specifically designed to expose catalyst materials to a slipstream of real flue gas over an extended period of time. The test unit is small enough to readily be moved from site to site. It is contained within a heated box that is mounted directly to a flue gas duct. Flue gas is continuously withdrawn from the duct through a heated glass probe and passes through a quartz filter before contacting the catalyst beds. Although the catalyst materials would be exposed to fly ash in future full-scale systems, fly ash is removed in the test unit to prevent plugging and "background" oxidation. The gas sample probe is oversized to minimize the collection of fly ash with the gas sample, and thus the amount of fly ash collected on the filter.

After passing through the filter, the flue gas passes through three parallel catalyst test cells. Each test cell contains two packed beds of catalyst material. The catalyst bed temperatures are controlled by adjusting the temperature of the heated box. Orifice plates and differential pressure transmitters are used to monitor the gas flow rate through each test cell. The gas flow rates are adjusted with a needle valve. After flowing through the cells, the flue gas passes through a condenser to remove moisture then through sample pumps. The pumps are able to pull up to 30 L/min of flue gas. With 7 L/min of flue gas passing through a test cell at 300°F, the nominal gas velocity is about 18 ft/min, which is similar to the gas velocity through a fabric filter in the EPRI COHPAC configuration (i.e., a pulse-jet fabric filter installed downstream of an ESP).

Since there are two catalyst beds in each of the three test cells, up to six different catalyst materials can potentially be tested simultaneously. Alternatively, two beds containing the same catalyst material can be tested in series to provide oxidation data at two space velocities (i.e., different ratios of volumetric flow rate to catalyst volume). The catalyst materials are mixed with sand before being placed in the test cells. The sand helps to minimize gas flow distribution problems by providing a thicker bed and more consistent pressure drop. Test ports are located at

the outlet of each catalyst bed as well as at the inlet and outlet of the quartz filter, so mercury samples can be collected at the inlet and outlet of all catalyst beds. By measuring the change in elemental mercury concentration across each bed, the level of oxidation can be determined.

Mercury measurements are made using a semi-continuous mercury analyzer developed for EPRI. As shown in Figure 1, flue gas is pulled from the catalyst test unit at about 1 L/min through a teflon-lined pump and passed through a series of impinger solutions. To measure total mercury in the flue gas, these impinger solutions consist of stannous chloride (SnCl₂) followed by a sodium carbonate (Na₂CO₃) buffer. The SnCl₂ solution reduces all flue gas mercury species to elemental mercury. After passing through the SnCl₂ impinger, the gas flows through the Na₂CO₃ solution to remove acid gases, thus protecting the downstream, analytical gold surface. Gas exiting the impinger solutions flows through a gold amalgamation column where the mercury in the gas is adsorbed at less than 100°C. After adsorbing mercury onto the gold for a fixed period of time (typically 10 minutes), the mercury concentrated on the gold is thermally desorbed (>700°C) from the column into nitrogen and sent as a concentrated stream to a cold-vapor atomic absorption spectrophotometer (CVAAS) for analysis. Therefore, the total flue gas mercury concentration is measured semi-continuously with a 10-minute sample time followed by a 10-minute analytical period.

To measure elemental mercury in the flue gas, the stannous chloride impinger is replaced with an impinger containing tris(hydroxymethyl)aminomethane (Tris) solution. The Tris solution has been shown in other EPRI studies to capture oxidized mercury while allowing elemental mercury to pass through without being altered¹. Mercury passing through the Tris solution to the gold is analyzed as described above and assumed to be elemental mercury only. The difference between the total mercury concentration (stannous chloride solution) and elemental mercury concentration (Tris solution) is the oxidized mercury concentration.

Using the equipment described above, the most promising catalyst materials will be tested for six months at each of three full-scale utility power plants. At each site, an initial set of short-term screening tests will be conducted to determine the best catalyst candidates at each site and to allow comparing the performance of different catalyst materials from site to site. Based on the results of these initial screening tests, four catalyst materials will be selected for long-term testing at each site. The materials will be placed in the catalyst test unit and exposed to flue gas continuously over a six-month period. Every four to six weeks, performance measurements will be taken to determine if oxidation has changed with time. Between performance measurements, the test unit operating conditions will be monitored remotely using mobile communications. This approach allows catalyst life to be estimated at three different coal-fired facilities.

The test sites will be chosen to provide a wide range of flue gas compositions and coal types. Testing has begun at the first site, which is a large (>500 MW), lignite-fired power plant. The second and third sites will likely be fired with subbituminous and a bituminous coals, respectively.

Laboratory tests will also be conducted to support the field work. The laboratory bench-scale unit shown in Figure 2 was used during Phase I, and will also be used in Phase II. The general test

approach consists of passing a simulated flue gas containing elemental mercury across a fixed-bed reactor that contains catalyst material. The gas exiting the fixed bed is analyzed semi-continuously by the same analytical technique described above to determine the fraction of inlet elemental mercury oxidized across the bed.

The simulated flue gas is prepared using reagent gases and calibrated flow meters. Elemental mercury is added to the simulated flue gas by passing nitrogen carrier gas across a mercury diffusion cell that contains a Hg^0 permeation tube. The amount of diffused mercury is controlled with the flow of nitrogen through the diffusion cell and the temperature of the diffusion cell. The mercury-containing nitrogen is then mixed with other flue gas components (SO_2 , HCl, NO_x , O_2 , CO_2 , and H_2O) at constant temperature before the gas enters the fixed-bed reactor.

The fixed-bed reactor consists of a mixture of catalyst material and sand placed in a temperature-controlled, vertical Pyrex column, typically yielding a bed length of about 1.75 inches. Gas exiting the fixed bed is analyzed to determine the percentage of inlet elemental mercury oxidized across the bed. The gas rate is typically about 1.4 L/min at 300°F, which results in a superficial gas velocity through the bed of about 30 ft/min.

The bench-scale unit will be used in Phase II to simulate the flue gas conditions of each full-scale test site before actually going to the field. This serves two purposes. First, information will be obtained about the expected effects of flue gas composition on oxidation before beginning the field tests. Second, field results can be compared to lab results to determine the effectiveness of the laboratory test unit as a screening device for predicting catalyst performance. The bench-scale unit may also be used develop a better understanding of oxidation mechanisms, to determine the activity of regenerated spent catalysts, and to better understand deactivation mechanisms, if necessary.

Results

Phase II results to date include laboratory results and initial catalyst screening test results from Site 1. Site 1 is a large (>500 MW) lignite-fired power plant. It was selected due to its relatively high concentration of elemental mercury and due to successful results at that site during Phase I. Table 1 summarizes the flue gas conditions at Site 1 as measured during Phase II in May 1998. Results from measurements collected in Phase I are included for comparison. Generally, the flue gas composition is consistent between Phase I and Phase II. However, the inlet mercury concentration varied over a wider range in 1998 than in 1997. During the initial Phase II tests, the total mercury concentration varied from 7 μ g/Nm³ to 22 μ g/Nm³. Similarly, the elemental mercury concentration varied from 4 to 16 μ g/Nm³. The inlet mercury oxidation percentage varied from 25% to 55%.

Table 2 summarizes the catalyst materials and conditions tested during the initial Site 1 catalyst screening tests. These materials were tested by mixing the indicated mass of material with 75 grams of sand and placing the mixture into the test cells. Some catalysts were placed in two

packed beds in series while others were placed in only one packed bed. Catalyst materials with more than one mass reported in Table 2 were tested as two beds in series. One of the six packed beds in the apparatus contained only sand to serve as a blank.

The catalyst samples in Table 2 can be classified as carbon-based materials, palladium-based materials, or fly ashes. Samples Carbon #1 and Carbon #2 are commercially available activated carbons. The other carbon-based materials are experimental materials derived from various lower-cost carbon sources. The palladium-based materials are commercially available. The fly ash samples were obtained from various coal-fired power plants.

Table 3 summarizes the initial catalyst screening test results obtained at Site 1 and compares them to results obtained in the lab at a similar flue gas composition. Two sand blank measurements were made which resulted in an average oxidation of 5.8%. This indicates that the sand does not significantly affect oxidation. In addition, the total mercury concentrations at the inlet and outlet of the catalyst beds were essentially equal during each test except when testing catalysts Carbon #6 and Pd #1. The lower outlet mercury concentrations obtained during these tests most likely indicate that mercury was being adsorbed by the catalyst material when the measurements were taken. During all of the other tests, adsorption equilibrium was apparently achieved before taking oxidation measurements, as indicated by the essentially equal inlet and outlet mercury concentrations.

All of the carbon-based catalysts achieved greater than 90% elemental mercury oxidation at Site 1 with the exception of catalyst Carbon #5. This material showed good oxidation in the lab but not in the field. The palladium-based catalysts also resulted in reasonably high levels of oxidation, but their performance was lower than that of the carbon-based materials. The fly ash samples were only able to achieve about 60% oxidation with the exception of SBA #5. This ash is known to have a high loss on ignition. These data suggest that the unburned carbon content in the fly ash affects mercury oxidation, since its performance was identical to that of the carbon-based materials.

In general, the lab results are reasonably consistent with the field results, although for three samples, Carbon #5, BA #3, and SBA #4, the lab result was significantly higher than the field result. Lab data will be compared to field data throughout the program to determine if the lab test procedure provides a good screening tool for catalyst performance.

Based on the initial screening test results, the following materials were selected for long-term testing at Site 1: Carbon #1, Carbon #4, Carbon #6, and SBA #5. Table 4 illustrates how these materials were configured in the catalyst test unit. The heated box temperature was set at 300°F, and the flow rate through each test cell was set at about 5 L/min at ambient temperature. Oxidation measurements across the beds will be repeated after about 6 weeks of flue gas exposure.

Future Activities

Oxidation measurements at Site 1 will be repeated every 4 to 6 weeks to determine any changes in oxidation percentages with time. If a catalyst material stops oxidizing elemental mercury, the catalyst will either be replaced with a larger mass of the same material or replaced with an entirely new catalyst. Any deactivated catalyst will be investigated to determine what caused the loss of activity.

Testing will continue at Site 1 for about five more months before moving to Site 2. A list of other potential test sites has been developed, and these sites will be contacted to determine interest in participating. One site will likely fire a Wyoming subbituminous coal, and one will fire a Kentucky or West Virginia bituminous coal. These sites would represent the greatest number of megawatts of U.S. power generation with installed FGD systems.

There are also plans to prepare one or more of the most promising catalysts in a configuration that could be used in future commercial applications (e.g., a honeycomb configuration). Once prepared, these samples will also be tested on flue gas from one or more of the test sites.

Acknowledgments

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References

1. Carey, T.R.; Hargrove, O.W., Jr.; Seeger, D.M.; Richardson, C.F.; Rhudy, R.G.; Meserole, F.B. Presented at the AIChE Spring National Meeting, Session 47, New Orleans, LA, February 25-29, 1996.

Table 1. Average Flue Gas Conditions for Site 1 Tests

Parameter	1998 Phase II Average	1997 Phase I Average
Inlet [Hg ⁰]	9 μg/Nm ³	$10 \mu\mathrm{g/Nm}^3$
Inlet [Hg total]	15 μg/Nm ³	$13 \mu\mathrm{g/Nm}^3$
SO_2	1500 ppm	1400 ppm
SO_3	0.7 ppm	-
HC1	1.8 ppm	1 ppm
NO_X	150 ppm	190 ppm

Table 2. Catalyst Materials and Conditions Tested at Site ${\bf 1}$

		Mass		Gas Rate
Catalyst		Tested	Temp.	(L/min @
Samples	Description	(mg)	(°F)	85°F)
Carbon #1	Carbon-based	75, 150	300	5
Carbon #2	Carbon-based	75, 150	300	5
Carbon #3	Carbon-based	75	300	5
Carbon #4	Carbon-based	75, 150	300	5
Carbon #5	Carbon-based	75, 150	300	5
Carbon #6	Carbon-based	75, 150	300	5
Pd #1	Palladium-based	750, 1500	300	5
Pd #3	Palladium-based	750	300	5
BA #2b	Bituminous ash	1500	300	5
BA #3	Bituminous ash	750, 1500	300	5
SBA #4	Subbituminous ash	1500	300	5
SBA #5	Subbituminous ash	750, 1500	300	5

Table 3. Initial Catalyst Performance Results at Site 1

	Site 1 Inlet		Site 1 Outlet		Hg ⁰ Oxidized by Catalyst			
	Total Hg	Hg^{0}	%	Total Hg	Hg^{0}	%	Site 1 Result	Lab Result
Sorbent	$(\mu g/Nm^3)$	$(\mu g/Nm^3)$	Oxidized	$(\mu g/Nm^3)$	$(\mu g/Nm^3)$	Oxidized	(%)	(%)
Sand	9.8	7.3	25.5	10.1	6.7	33.7	8.2	-
Sand	18.1	9.0	50.3	17.5	8.7	50.3	3.3	3
Carbon #1	9.8	7.3	25.5	10.6	0.0	100.0	100.0	100
Carbon #2	10.2	7.5	26.5	10.6	0.3	97.6	96.7	96
Carbon #3	10.2	7.5	26.5	11.0	0.5	95.5	93.3	91
Carbon #4	7.0	3.7	47.1	6.5	0.2	96.9	94.6	-
Carbon #5	8.8	4.1	53.4	7.9	1.6	79.7	61.0	100
Carbon #6	18.1	9.0	50.3	13.6	0.1	99.3	98.9	97
Pd #1	19.0	9.8	48.4	14.6	1.0	93.5	90.3	91
Pd #3	18.1	9.0	50.3	19.9	1.2	94.0	86.7	98
BA #2b	26.1	15.3	41.4	23.0	6.7	70.9	56.2	49
BA #3	21.5	16.2	24.7	23.3	6.3	73.0	61.1	96
SBA #4	8.8	4.1	53.4	6.6	1.7	74.2	58.5	97
SBA #5	21.5	16.2	24.7	19.7	0.0	100.0	100.0	-

Table 4. Catalyst Configuration for Long-Term Testing at Site 1

	Test Cell #1	Test Cell #2	Test Cell #3
Packed Bed #1	75 g of sand	0.25 g Carbon #1 in 75 g sand	0.25 g Carbon #6 in 75 g sand
Packed Bed #2	1.5 g SBA #5 in 75 g sand	0.25 g Carbon #1 in 75 g sand	0.25 g Carbon #4 in 75 g sand

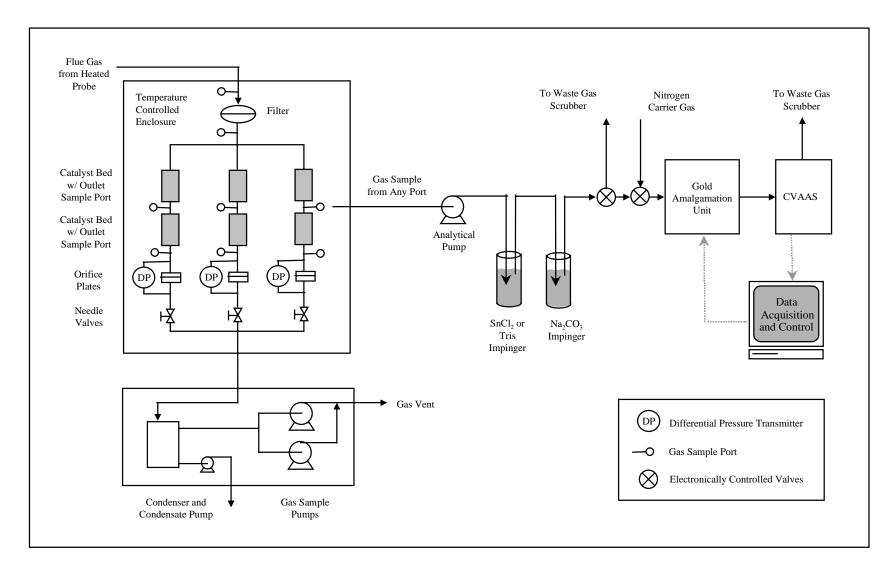


Figure 1. Catalyst Field Test Unit and Mercury Analyzer System

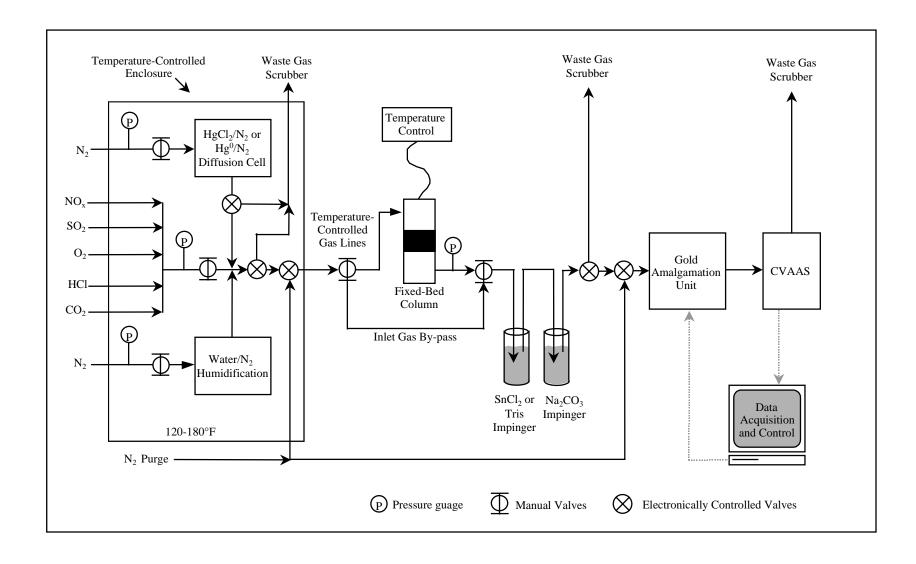


Figure 2. Bench-Scale, Fixed-Bed Mercury Oxidation Test Unit

Enhanced Control of Mercury by Innovative Modifications to Wet FGD Processes

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Background

- 1990 Clean Air Act amendments mandated a study of the effect of Hg emissions from power plants.
- Hg is emitted in extremely low concentrations, but mostly in the vapor phase.
- Hg is not effectively removed in particulate control devices.

Background (Continued)

- Oxidized forms of Hg are removed in wet scrubbers, but Hg⁰ is not.
- Approximately 5% to 75% of the Hg in flue gas from coal-fired power plants is in the form of Hg⁰.

Project Description

- Part of the Mega-PRDA program; being conducted in two phases.
- Phase I was completed in September 1997.
- Phase I investigated three concepts for enhanced HAPs removal.
- Of those, catalytic oxidation of Hg⁰ to improve removal in wet FGD appeared most promising.

Project Description (Continued)

- Phase I identified catalysts capable of achieving up to 100% oxidation of Hg⁰.
- Phase I included proof-of-concept testing at laboratory and pilot scale with synthetic, coaland lignite-fired flue gases.
- Phase II will determine catalyst life in slipstream testing at three coal-fired power plants.
- Phase II will also be supported by laboratoryscale investigations.

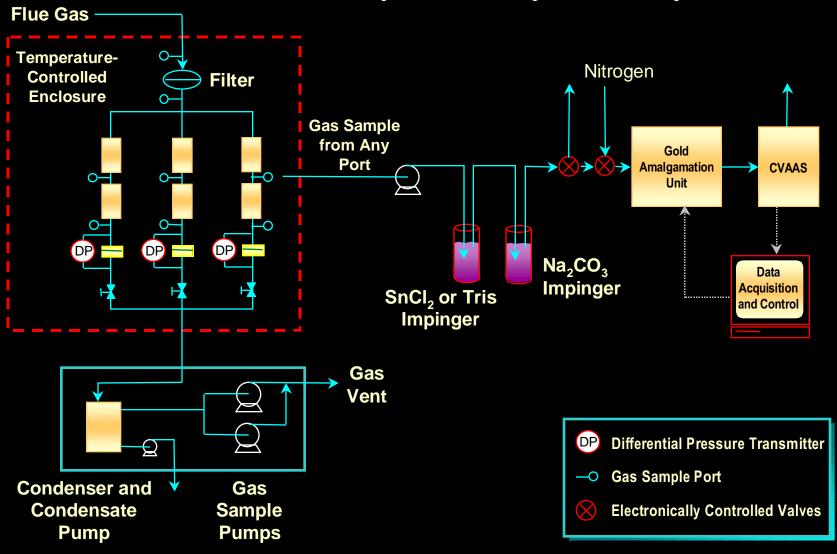
Phase II Objectives

- Determine the effectiveness of catalysts identified in Phase I at several coal-fired sites.
- Estimate catalyst life in various flue gas streams.
- Determine the volume of catalyst required to achieve at least 70% oxidation of Hg⁰ over an extended period of time.

Phase II Approach

- Use a transportable slipstream test unit to allow long-term testing of three or more catalysts at a time.
- Conduct long-term testing at three sites that are representative of the range of coals that are fired at power plants with FGD.

Catalyst Field Test Unit and Mercury Analyzer System



Potential Coal Effects on Hg Oxidation

- Coal chloride appears to promote oxidation.
- Flue gas SO₃ content may deactivate some catalysts.
- Boiler type, NO_X levels, and fly ash composition also appear to influence oxidation.

Coals Represented at Full-Scale Test Sites

- Texas Lignite (8% of FGD MW)
- Wyoming (PRB) Subbituminous (15%)
- Kentucky (18%) or West Virginia (10%)
 Bituminous

Phase II Schedule

- Began in April 1998.
- Catalyst screening tests at first site in May 1998.
- Catalyst life tests at first site began June 1998.
- Testing at the second site will begin about the end of 1998.

Initial Results – Site 1

- Total Hg concentrations vary from 7 to 22 μg/Nm³.
- Hg⁰ concentrations vary from 4 to 16 μg/Nm³.
- Inlet Hg oxidation varies from 25% to 55%.

Average Flue Gas Conditions for Site 1 Tests

Parameter	Average
Inlet [Hg ⁰]	9 μg/Nm³
Inlet [Hg total]	15 μg/Nm ³
SO_2	1500 ppm
SO_3	0.7 ppm
HCI	1.8 ppm
NO_x	150 ppm
H_2O	12% - 13%

Catalyst Materials and Conditions Tested at Site 1

Catalyst Samples	Description	Mass Tested (mg)
Carbon #1 Carbon #2 Carbon #3 Carbon #4 Carbon #5 Carbon #6	Carbon-based Carbon-based Carbon-based Carbon-based Carbon-based Carbon-based	75, 150 75, 150 75 75, 150 75, 150 75, 150
Pd #1 Pd #2	Palladium-based Palladium-based	750, 1500 750
BA #2b BA #3 SBA #4 SBA #5	Bituminous ash Bituminous ash Subbituminous ash Subbituminous ash	1500 750, 1500 1500 750, 1500

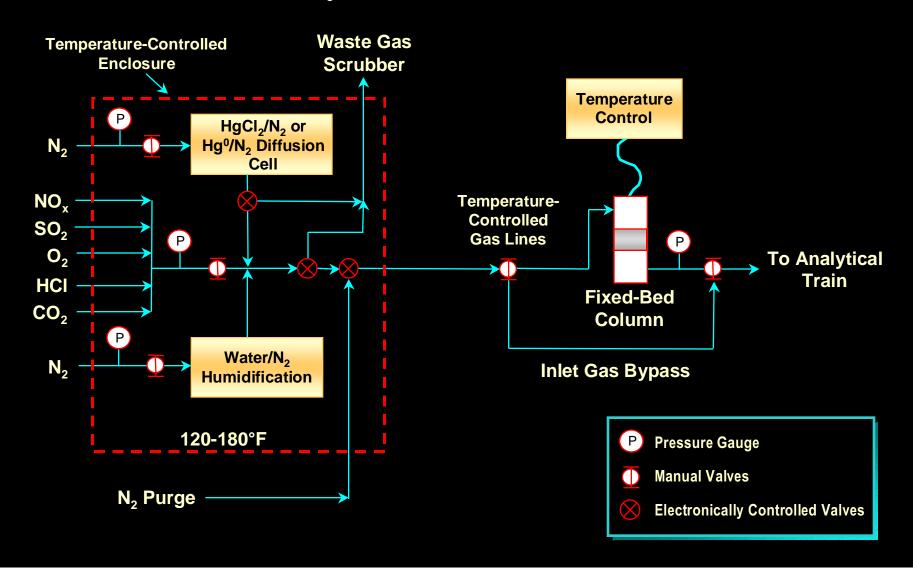
Initial Catalyst Performance Results at Site 1

Hg Oxidation at Bed Inlet	Hg Oxidation at Bed Outlet	Hg ⁰ Oxidation Across Bed (%)
26	34	8
50	50	3
26	100	100
27	98	97
27	96	93
47	97	95
53	80	61
50	99	99
48	94	90
50	94	87
41	71	56
24	73	61
53	74	59
25	100	100
	26 50 26 27 27 47 53 50 48 50 41 24 53	at Bed Inlet at Bed Outlet 26 34 50 50 26 100 27 98 27 96 47 97 53 80 50 99 48 94 50 94 41 71 24 73 53 74

Comparison of Site 1 Results to Laboratory Results at Similar Conditions

Sorbent	Hg ⁰ Oxidation Across Bed (%)	Lab Result (%)
Sand	8	3
Sand	3	3
Carbon #1	100	100
Carbon #2	97	96
Carbon #3	93	91
Carbon #4	95	
Carbon #5	61	100
Carbon #6	99	97
Pd #1	90	91
Pd #3	87	98
BA #2b	56	49
BA #3	61	96
SBA #4	59	97
SBA #5	100	

Bench-Scale, Fixed-Bed Mercury Oxidation Test Unit



Catalyst Configuration for Long-Term Testing at Site 1

	Test	Test	Test
	Cell #1	Cell #2	Cell #3
Packed	75 g Sand	0.25 g	0.25 g
Bed #1		Carbon #1	Carbon #6
Packed	1.5 g SBA #5	0.25 g	0.25 g
Bed #2		Carbon #1	Carbon #4

All beds operate at 300°F, 5 L/min (85°F) gas flow.

Summary

- Phase I showed that a number of substances can catalyze the oxidation of Hg⁰ in flue gas to a form that can be removed in a wet scrubber.
- Phase II will determine catalyst volume requirements and life in three actual coal-fired flue gases.
- Catalyst screening at Site 1 confirmed Phase I results; catalyst life tests at Site 1 have just begun.